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Substrate melting during laser heating of nanoscale metal films

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ABSTRACT

We investigate heat transfer mechanisms relevant to metal films of nanoscale thickness deposited on a silicon (Si) substrate coated by a silicon oxide (SiO_2) layer and exposed to laser irradiation. Such a setup is commonly used in the experiments exploring self and directed assembly of metal films that melt when irradiated by laser and then evolve on time scale measured in nanoseconds. We show that in a common experimental setting, not only the metal but also the SiO_2 layer may melt. Our study of the effect of the laser parameters, including energy density and pulse duration, shows that melting of the substrate occurs on spatial and temporal scales that are of experimental relevance. Furthermore, we discuss how the thicknesses of metal and of the substrate itself influence the maximum depth and liquid lifetime of the melted SiO_2 layer. In particular, we find that there is a minimum thickness of SiO_2 layer for which this layer melts and furthermore, the melting occurs only for metal films of thickness in a specified range. In the experiments, substrate melting is of practical importance since it may significantly modify the evolution of the deposited nanoscale metal films or other geometries on nanoscale.

1. Introduction

Nanoparticle self-assembly, induced by dewetting of thin nanoscale metallic films, has recently been shown to be a promising process for synthesizing large-area nanoparticle ensembles, see Ref. [1] for a recent review. The process involves nanosecond pulsed-laser-induced melting of metallic films, filaments, and other geometries, followed by dewetting that may lead to formation of patterns characterized by a particular spatial order and size distribution [2–8]. Due to its relevance to a number of applications involving metal particles on nanoscale, the pulsed-laser dewetting has recently been studied in great detail [9–21]. While these studies lay out a clear picture of the basic physical mechanisms of the dewetting, understanding of relevant thermal phenomena is still limited, and there are only few studies discussing these effects [4,10,22,12,23]. In particular, to the authors' knowledge, not much attention has been paid to the thermal processes in the substrate itself, leading possibly to its melting. As an example motivating such a study, Fig. 1 shows an experimental image from Ref. [11] that suggests that high temperatures reached during laser irradiation may lead to partial melting of the substrate. This figure shows that after the initial laser pulse, the liquid metal filament recesses into the substrate, suggesting substrate melting. In such a scenario, the dewetting dynamics of the metal film can be strongly coupled to the substrate behavior, in particular since the dewetting of a metal film resting on a liquid substrate can be altered significantly. Therefore, there is a strong motivation to understand possible phase change of the substrate that may occur during experiments involving pulsed-laser irradiation of metal films and other geometries.

In this work we present a model describing the heat transfer and phase change of the metal/substrate setup during laser irradiation. While for definitiveness we use the material parameters that correspond to a three layer setup consisting of copper (Cu)/silicon dioxide (SiO_2)/ silicon(Si) (see Fig. 2), we will vary appropriate nondimensional parameters in order to reach a more general understanding of the problem. Regarding Cu/SiO_2/Si setup, we note that while SiO_2 may be native to the Si substrate, we concentrate here on a common setup such that an SiO_2 layer of controlled thickness (typically 100 nm) is applied to the Si substrate, see e.g. Ref. [24]. The laser irradiation is considered to be applied via a Gaussian beam, and we study the effect of the laser parameters, including energy density and pulse duration, on possible melting of the SiO_2 layer. For the parameters for which the SiO_2 layer melts, we compute the depth of the melted SiO_2 layer and the total time the layer remains in liquid phase. We find that maximum thickness of the melted region can be of the same order of magnitude as the thickness of the metal film. Finally, we study the effect of changing

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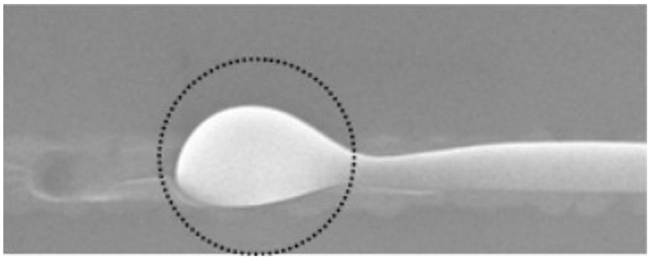

Fig. 1. Experimental image from Ref. [11]. After the initial laser pulse, the liquid metal filament (nickel) recesses slightly into the substrate (Si with a SiO_2 layer atop), suggesting substrate melting.

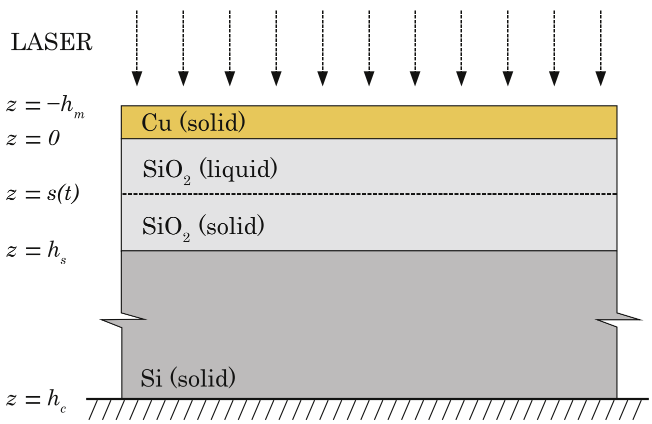

Fig. 2. A schematic of the $Cu/SiO_2/Si$ thin film structure, assuming that SiO_2 layer is partially melted; the melting front is represented by the dashed line in the SiO_2 layer (not to scale).

the metal and substrate thickness on the maximum depth and liquid lifetime of the melted SiO_2 layer.

The rest of the paper is organized as follows. We begin in Section 2 by presenting the governing equations. In Section 3, we present the results, starting with the reference case (Section 3.1), followed by a discussion of the effects of key dimensionless variables, such as the ratio of the heat diffusion time to the laser pulse duration, and the ratio of energy per unit volume needed to melt the substrate to the energy per unit volume absorbed in the metal (Section 3.2). The role that the metal and substrate thicknesses play in melting process is discussed in Section 3.3. Section 4 summarizes the main conclusions.

2. Mathematical model

Fig. 2 illustrates the considered setup. A metal thin film of the thickness h_m is placed on top of a flat substrate of thickness h_c. The substrate consists of two layers - top layer of thickness h_s (SiO_2) that in turn lies on top of another layer of thickness $h_c - h_s$ (Si). The three-layer configuration, initially at room temperature, T_{room}, is irradiated by a Gaussian temporal laser pulse. Although our model is general and can be applied to any material, the $Cu/SiO_2/Si$ experimental setup reported elsewhere, see e.g. Ref. [25], will be taken as a *reference case*. Thus, we will make references to Cu, SiO_2 and Si layers throughout the paper to indicate metal, substrate 1 and substrate 2 layers, respectively.

To keep the formulation of the problem tractable and as general as possible, we make a number of assumptions and approximations that are listed next. Future work should consider relaxing some of these, particularly if direct comparison to experiments is attempted. The assumptions are as follows: (i) the substrate is completely transparent to the laser radiation [26], so that the metal film absorbs the energy of the laser and transfers it to the substrate by conduction; (ii) we will consider static setup for both metal and the substrate, and therefore convection is not considered in modeling heat transfer (this assumption is justified a posteriori in Section 3.1); (iii) only heat transfer in the out-of-plane (z) direction is considered, and (iv) the material properties are taken as temperature independent. For simplicity, for the metal film, we use the material parameters at ambient conditions; future works should consider the influence of the change of material properties with temperature on the results presented here; for reference, we note that, e.g., metal thermal conductivity changes about 15% between ambient and melting temperatures. Furthermore, we note that we focus on melting of the substrate itself, and do not discuss melting of the metal film that has been already considered in existing works [4,12,17]. These have also shown that the influence of phase change of metal on its temperature evolution is minor. Similar studies have also shown that laser energy typically used in experiments, ablation/evaporation of the metal film is negligible [27,28]. For instance, for the reference case considered in this study the mass loss was estimated to be <2% per laser pulse [28]. Therefore, thinning effects due to mass loss are not considered in the present manuscript. Finally, as we will discuss, the temperatures achieved at the Cu/SiO_2 interface will induce melting in the SiO_2 layer but the melt front will not reach the Si layer below for the SiO_2 thicknesses considered. Thus, we will focus on the configuration Cu/liquid-SiO_2/solid-SiO_2/solid-Si, as depicted in Fig. 2.

To describe the heating and possible melting of the substrate, we consider heat conduction in each layer coupled by appropriate boundary and interface conditions. The laser-metal interaction is introduced by the means of a source term in the heat equation for the metal layer [29].

Under the above assumptions, the governing equations for the temperature are as follows:

$$\rho_m c_m \frac{\partial T_m}{\partial t} = k_m \frac{\partial^2 T_m}{\partial z^2} + Q, \quad -h_m < z < 0, \tag{1}$$

$$\rho_l c_l \frac{\partial T_l}{\partial t} = k_l \frac{\partial^2 T_l}{\partial z^2}, \quad 0 < z < s(t), \tag{2}$$

$$\rho_s c_s \frac{\partial T_s}{\partial t} = k_s \frac{\partial^2 T_s}{\partial z^2}, \quad s(t) < z < h_s, \tag{3}$$

$$\rho_c c_c \frac{\partial T_c}{\partial t} = k_c \frac{\partial^2 T_c}{\partial z^2}, \quad h_s < z < h_c. \tag{4}$$

where T is the temperature, k the thermal conductivity, ρ the density and c the specific heat. The subscripts m, l, s, c indicate Cu, liquid-SiO_2, solid-SiO_2 and Si, respectively. The phase change front is represented by $s(t)$.

The laser beam is applied in the z direction. It is of the form [29]

$$Q = E_0(1-R)g(x,y)f(z)q(t), \tag{5}$$

where E_0 is the laser energy density and R is the reflectivity of the Cu film. The attenuation within the metal is described by Beer's law, $f(z) = \alpha e^{-\alpha z}$, where α is the absorption coefficient [29]. The pulse is taken as Gaussian, and is specified by $q(t) = (1/\sigma\sqrt{2\pi})\exp\left(-(t-t_p)^2/2\sigma^2\right)$. We choose the standard deviation, $\sigma = t_p/2\sqrt{2\ln(2)}$, giving the full width at half maximum equal to t_p.

The term $g(x,y)$ accounts for possible spatial variation of the pulse. For metals on nanoscale, the spatial extend of a laser spot is much larger than any other relevant lenthscale, motivating consideration of a uniform pulse, leading to $g(x,y) = 1$. Therefore,

$$Q = \frac{2\sqrt{\ln(2)}(1-R)E_0\alpha}{t_p\sqrt{\pi}} e^{-4\ln(2)\left(\frac{t}{t_p}-1\right)^2-\alpha(z+h_m)}, \tag{6}$$